

POPULAR SUMMARY

Retrieval of aerosol optical depth above clouds from OMI observations: sensitivity analysis, case studies

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Aerosols are tiny particles, such as smoke, soil dust, and sea salt, suspended in the atmosphere. They scatter and absorb solar radiation and play an important role in the energy balance of the earth-atmosphere system. Carbonaceous particles produced by biomass burning and boreal forest fires, and desert dust particulate originated in the arid and semi-arid regions are the most predominant light absorbing aerosol types. The UV Aerosol index detects the presence of absorbing aerosols. A large fraction of the atmospheric aerosol load reaches the free troposphere and is frequently located above clouds.

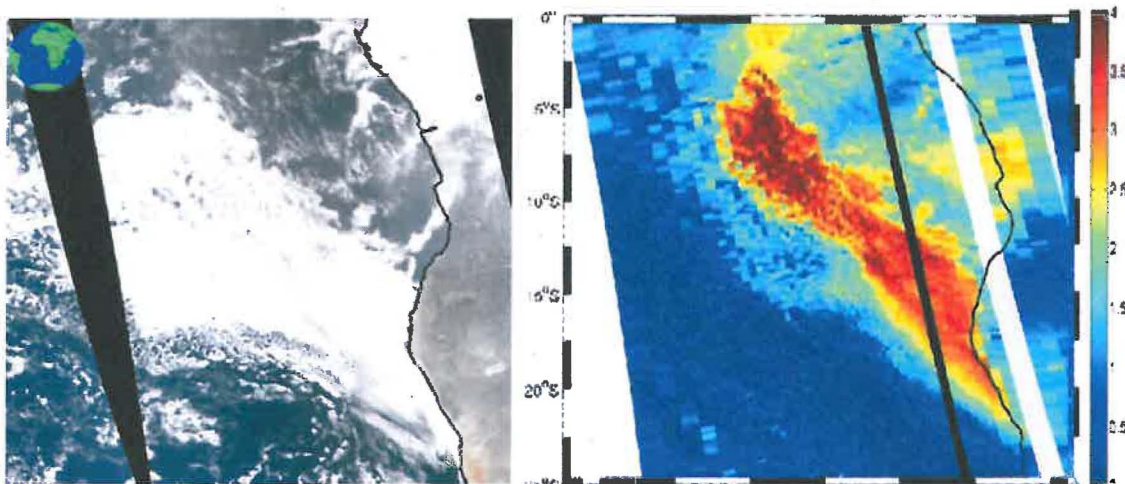


Figure 1: An absorbing aerosol layer as seen from space by two satellite sensors on August 4, 2007 over the South Atlantic Ocean off the coasts of Angola and Namibia : Aqua-MODIS visible image of an extended low level cloud (left); Aura-OMI detection of a smoke layer above clouds in terms of the UV aerosol index.

A variety of well established space and ground based techniques are used to measure the amount (i.e., optical depth) of suspended particle matter under cloud-free conditions. Aerosol above clouds, however, can only be measured from airborne or satellite-based instrumentation.

We have developed a method to measure the optical depth of light-absorbing smoke and dust aerosols located above clouds using satellite observations of upwelling radiation at two wavelengths in the near-UV spectral range (330-390 nm) used to calculate the UV Aerosol Index. The technique takes advantage of the particle's large absorption capacity of cloud reflected radiation to detect and quantify particulate matter above clouds.

Carbonaceous and dust aerosols above clouds absorb a fraction of the radiation reflected by the cloud and can, thus, produce a net warming effect of the atmospheric column. The magnitude of the resulting warming depends on the aerosol optical depth and its absorption capacity. The measuring technique developed here provides a very important remote sensing tool to accurately quantify the atmospheric aerosol load above clouds. The improved AOD estimates will allow a better characterization

the direct radiative forcing of above-cloud-aerosols, and a better understanding of other aspects of aerosol-cloud interaction.

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18 **Retrieval of Aerosol Optical Depth above Clouds from OMI Observations:**
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Abstract

A large fraction of the atmospheric aerosol load reaching the free troposphere is frequently located above low clouds. Most commonly observed aerosols above clouds are carbonaceous particles generally associated with biomass burning and boreal forest fires, and mineral aerosols originated in arid and semi-arid regions and transported across large distances, often above clouds. Because these aerosols absorb solar radiation, their role in the radiative transfer balance of the earth atmosphere system is especially important. The generally negative (cooling) top of the atmosphere direct effect of absorbing aerosols, may turn into warming when the light-absorbing particles are located above clouds. The actual effect depends on the aerosol load and the single scattering albedo, and on the geometric cloud fraction. In spite of its potential significance, the role of aerosols above clouds is not adequately accounted for in the assessment of aerosol radiative forcing effects due to the lack of measurements. In this paper we discuss the basis of a simple technique that uses near-UV observations to simultaneously derive the optical depth of both the aerosol layer and the underlying cloud for overcast conditions. The two-parameter retrieval method described here makes use of the UV aerosol index and reflectance measurements at 388 nm. A detailed sensitivity analysis indicates that the measured radiances depend mainly on the aerosol absorption exponent and aerosol-cloud separation. The technique was applied to above-cloud aerosol events over the Southern Atlantic Ocean yielding realistic results as indicated by indirect evaluation methods. An error analysis indicates that for typical overcast cloudy conditions and aerosol loads, the aerosol optical depth can be retrieved with an accuracy of approximately 54% whereas the cloud optical depth can be derived within 17% of the true value.

1. Introduction

Because of the nature of the physical processes driving the emission and atmospheric injection of desert dust and carbonaceous aerosols, i.e., the wind's lifting power of soil particles over arid and semi-arid areas and the strong convective activity associated with anthropogenic biomass burning and wild fire events, large amounts of these light-absorbing particles reach the free troposphere more often than other aerosol types that generally reside in the boundary layer such as sulfate and sea salt aerosols. Elevated layers of desert dust and carbonaceous particulate are therefore frequently observed above clouds where they are mobilized by the prevailing winds and transported thousands of kilometers away from their original sources. The spring and summer transoceanic transport of desert dust across the Atlantic Ocean and the spring flow of aerosol from Asian sources across the Pacific Ocean to North America and sometimes reaching Northern Europe have been well documented using both satellite observations [Kaufman *et al.*, 2005], as well as model calculations [Kallos *et al.*, 2006; Johnson *et al.*, 2010]. Smoke layers originating from well-known regional sources of agricultural biomass burning mainly in Africa and South America [Duncan *et al.*, 2003; Torres *et al.*, 2010]. Smoke layers can also be the result of wild fires in both hemispheres [Colarco *et al.*, 2004; Dirksen *et al.*, 2009] that are transported across the oceans or from northern to mid-latitudes from their original distant remote sources to populated centers where they often contribute to the observed high levels of local pollution [Jaffe *et al.*, 2004]. Elevated aerosol layers, often lofted above clouds, are also observed in connection with pyro-convective events [Fromm *et al.*, 2008]. Much of the long range transport of desert

dust and carbonaceous aerosols, however, takes place above low level layers of altostratus and stratocumulus clouds.

Quantifying the fraction of the atmospheric aerosol load above clouds is important for air quality considerations as some of these aerosols eventually descend into the boundary layer contributing to the enhancement of pollution levels. Knowledge of the amount and type of aerosol above clouds is also of interest for radiative forcing considerations. The general cooling effect of these aerosol types under clear sky conditions may turn into a warming effect when located above clouds. Depending on their light absorption capacity above-cloud-aerosols may have a positive radiative forcing effect at the top of the atmosphere [Keil and Haywood, 2003; Haywood and Shine, 1997; Chand et al., 2008], whose magnitude depends on the aerosol single scattering albedo and cloud fraction in addition to the total aerosol load. Liao and Seinfeld [1998a, 1998b] examined the radiative forcing of desert dust and found out that the highest radiative forcing occurs for dust layers located above clouds. This effect is particularly important for carbonaceous aerosols in which the combined absorption effect of its organic (OC) and black (BC) carbon components acts on the incoming solar radiation from the UV to the near-IR producing potentially large effects on the local climate. The radiative effect of carbonaceous aerosols above stratiform clouds off the coasts of Angola and Namibia was estimated during the SAFARI 2000 field experiment making use of radiative transfer calculations [Keil and Haywood, 2003] and airborne-measured profiles of aerosol particle number concentration, scattering coefficients, and meteorological parameters [Haywood et al., 2003]. The study indicated that the normally cooling effect of the smoke aerosol layer of about -13 Wm^{-2} over a cloud free area turned into a warming effect of 11.5 Wm^{-2}

for the same aerosol layer located above a cloud deck which is similar to the expected radiative forcing effect associated with an increase in surface albedo. Changes in cloud morphology may also result as a consequence of atmospheric heating (by aerosol absorption) of the air above the cloud deck. As suggested by *Wilcox* [2010], this aerosol-induced heating of the atmosphere may lead to the optical thickening of the cloud resulting from enhanced liquid water path and lower cloud top associated with the warming above the cloud.

Agriculture-related biomass burning in the tropical regions accounts for nearly 80 % of the global amount including both anthropogenic and naturally produced burning [*Hao and Liu*, 1994]. It is estimated that although BC above clouds makes up 20% of the total burden, it accounts for about 50% of the BC radiative forcing effect [*Zarcichy and Bond*, 2010]. To properly estimate the regional and global forcing effect of carbonaceous and desert dust aerosols it is therefore necessary to quantify the atmospheric aerosol load above clouds.

In this paper we present a method of deriving aerosol optical depth over clouds making use of near-UV observations by the Ozone Monitoring Instrument (OMI) on the Aura satellite. The high near-UV sensitivity to aerosol absorption [*Torres et al.*, 1998; 2007] is further enhanced in the presence of underlying clouds due to the high reflectance from the cloud-top layer. A general description of the near-UV capability of aerosol detection is presented in section 2 followed by a detailed discussion of a sensitivity analysis of near-UV observations to aerosol presence above clouds in section 3. A description of the retrieval procedure and error analysis is presented in section 4, and

results of application of the inversion procedure to two case studies are discussed in section 5. Discussion and conclusion are presented in section 6.

2. Satellite detection of aerosols above clouds.

The detection and characterization of aerosols in cloud-free scenes using near-UV satellite observations is an established aerosol remote sensing technique developed based on Total Ozone Mapping Spectrometer (TOMS) observations [Torres *et al.*, 1998]. The near-UV technique is particularly sensitive to absorbing aerosols as explained below. A long-term record (1979-1992/1996-2001) of aerosol optical depth [Torres *et al.*, 2002] was developed from TOMS observations. Currently, OMI near-UV observations are used in the OMAERUV algorithm to derive aerosol optical depth (AOD) and single scattering albedo (SSA) under cloud-free conditions [Ahn *et al.*, 2008; Torres *et al.*, 2007]. In this paper we describe initial results of a recently developed approach using OMI observations to derive the optical depth of aerosol layers above clouds.

Aerosols above clouds, or for that matter above any bright background, are not easily detectable by conventional satellite remote sensing approaches that measure aerosols based on the increased reflectance associated with particle scattering. The difficulty lies on the fact that aerosol scattering contribution to the total upwelling reflectance at the top of the atmosphere is very small compared to the reflectance of the bright cloud background. Thus, to be able to separate the aerosol scattering signal from the measured total, one should be able to characterize the reflective properties of the bright background (i.e., clouds) to a level of accuracy probably unachievable. Aerosol

absorption effects, however, can be advantageously used for the detection and characterization of absorbing particles even over highly reflective backgrounds.

It has been demonstrated that aerosols that absorb UV radiation can be detected and characterized using the unambiguous particle absorption signal in the measured levels of UV reflectance [Torres *et al.*, 1998]. The unique advantage of the near-UV detection capability of absorbing aerosols is the large Rayleigh scattering component that makes available for absorption by aerosol particles a significantly larger number of photons than at longer wavelengths [Torres *et al.*, 1998]. The most commonly known near-UV aerosol product is the absorbing aerosol index (AAI), a parameter used to detect UV-absorbing aerosols over ocean and all land surfaces.

The AAI was developed as a byproduct of improvements to the column ozone amount retrieval algorithm applied to UV observations by the TOMS instrument. It is basically a residual quantity resulting from the comparison between measured and calculated radiances in the range 330-390 nm where trace gas absorption effects are negligible. The calculated radiance is obtained using a simple model of the earth-atmosphere system consisting of a molecular atmosphere bounded at the bottom by a Lambert Equivalent Reflector (LER) [Dave and Mateer, 1967]. The LER reflectivity ρ is derived from measurements at wavelength λ_0 . A key assumption in this model representation of the earth-atmosphere system is that the reflectivity of the column atmosphere's lower boundary is wavelength independent in the near UV. This hypothetical surface is intended to account for the wavelength dependent effects of surface, clouds, and aerosols which are not explicitly included in the radiative transfer calculations.

The residue, r_λ is defined as the natural log of the ratio of the actually measured radiance L_λ^* to the calculated value L_λ^{cal} assuming an effective surface reflectivity ρ_{λ_0} ,

$$r_\lambda = -100 \log \left[\frac{L_\lambda^*}{L_\lambda^{cal}(\rho_{\lambda_0})} \right] \quad (1).$$

This residual quantity provides a measure of the error in predicting the actually observed spectral contrast using the LER model. Examination of global maps of the residue clearly indicate that UV-absorbing aerosols are by far the most important residue source [Hsu *et al.*, 1996; Herman *et al.*, 1997] and has become a powerful tool of absorbing aerosols detection that has been used in a variety of applications. Hence, the term Absorbing Aerosol Index was coined to refer to this residual quantity. The AAI is the same as r_λ when $\lambda_0 > \lambda$, and it is defined as $-r_\lambda$ when $\lambda_0 < \lambda$ so that it is always positive in the presence of absorbing aerosols. In the OMAERUV algorithm AAI is calculated for $\lambda_0 = 388$ nm and $\lambda = 354$ nm.

The AAI detection capability of absorbing aerosols is not limited to cloud-free, dark scenes. It can also identify the presence of absorbing particles above bright backgrounds such as ice-snow covered surfaces and cloud decks. Figure 1 shows a typical aerosol-above-cloud event on August 4, 2007 over the Southern Atlantic Ocean as seen by three different A-train sensors. The top panel depicts the OMI UV Aerosol Index indicating the unmistakable presence of an absorbing aerosol layer above the horizontally extended cloud feature shown in the Aqua-MODIS (Moderate Resolution Imaging Spectro-radiometer) true color image composite (bottom panel). The cloud layer covers a large region from the coast of Namibia and Angola extending into the ocean in a north-west direction. The center panel illustrates the vertical distribution of clouds and aerosols

in terms of the CALIOP (Cloud Aerosol Lidar with Orthogonal Polarization) lidar 532 nm attenuated backscatter measured along the CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation) spacecraft orbital track shown on both the OMI and MODIS images. The CALIOP's transect shows the presence of a geometrically thin low level cloud with cloud-top at 1 km above sea level. An aerosol layer at about 2 km above the cloud is observed over the first half of the image. Towards the middle of the shown orbital section the aerosol layer is observed over a cloud free area.

TOMS and OMI AAI observations over clouds have been used in conjunction with other satellite measurements to estimate the radiative effects of aerosol layers above clouds [Hsu *et al.*, 2003; Peters *et al.*, 2009]. Other applications include the use of SCIAMACHY derived AAI for identification of aerosol layers above cloud and the analysis of aerosol absorption effect on visible and near-IR TOA radiance measurements by the same sensor [de Graaf *et al.*, 2007].

Aerosols above clouds have also been detected and their optical depth estimated by polarization measurements by the POLDER sensor [Waquet *et al.*, 2009]. This approach uses multi-sensors measurements aboard A-train constellation to derive aerosol optical depth above cloud taking advantage of the unique angular dependence associated with the presence of the polarized radiation of above-cloud-aerosols. The modification in the polarized light reflected by cloud top by the aerosol layer allows retrieving aerosol optical depth using a single scattering approximation for aerosol signal.

Active space-borne measurements can also be used to derive the optical depth of aerosols above clouds. The standard CALIOP AOD product is derived as the vertically integrated aerosol extinction coefficient over the vertical extent of the identified aerosol

feature. This approach requires an assumption of the extinction-to-backscatter or lidar ratio [Kittaka *et al*, 2011]. Alternate transmission methods for directly deriving AOD above clouds from CALIOP observations of depolarization [Hu *et al*, 2007] and color [Chand *et al*, 2008] ratios have been investigated. Unlike the standard approach, the transmission-based lidar techniques do not require assumption on the lidar ratio.

3.0 Sensitivity Analysis

3.1 Aerosol and Cloud Models

In this section we discuss the results of a sensitivity analysis of the near-UV reflectance at the top of the atmosphere to aerosol and cloud properties. Radiative transfer calculations at 354 and 388 nm were carried out for an atmosphere containing both aerosols and clouds using the Vector Linearized Discrete Ordinate Radiative Transfer code (VLIDORT) developed by Spurr [2006]. VLIDORT calculates Jacobians and radiances for reflectances at the top of a multilayered atmosphere. It takes into account the atmosphere's curvature making use of a pseudo-spherical approximation for solar beam attenuation and includes a sphericity correction for off-nadir viewing at large angles. The VLIDORT code accounts for multiple scattering effects by molecules, aerosols and clouds.

In the following discussion, the cloud droplet size distribution is represented by the C1 modified Gamma distribution suggested by Deirmendjian (1964) which is commonly used in clouds radiative transfer modeling [Rossow and Schiffer, 1999; Ahmad *et al.*, 2004]. The assumed distribution with a maximum radius of 15 μm yields a 6.0 μm radiatively effective radius. Two aerosol models representative of carbonaceous

aerosols are used in the calculations. The assumed bi-modal particle size distributions and complex refractive index of the selected aerosol models are based on multiyear AERONET (Aerosol Robotic Network) statistics of measurements at locations where carbonaceous aerosols are typically observed [Dubovik, *et al.*, 2002]. AERONET retrievals of aerosol particle size and optical properties use both direct sun and sky radiance measurements as input to an inversion algorithm [Dubovik and King, 2000]. The reported measurements are representative of the atmospheric column aerosol load and are therefore suitable for the analysis of space-borne remotely sensed data. The first model, representative of black carbon (BC), has a wavelength independent imaginary refractive index, while the second one assumes a wavelength dependent absorption refractive index associated with organic carbon (OC) consistent with observations [Kirchstetter *et al.*, 2004]. Table 1 lists the aerosol parameters used in the calculations.

A 0.5 km thick cloud located between 1 and 1.5 km above sea level (900 and 850 hPa respectively) is assumed. The aerosol vertical distribution is represented by a single layer whose concentration follows a Gaussian distribution characterized by the height of maximum concentration (peak) and full-width-half-maximum. Note that this yields an aerosol profile in which the entire aerosol load is located above the cloud. The above described vertical distribution of clouds and aerosols resembles actual conditions observed by the CALIOP lidar over the Southern Atlantic Ocean as shown in Figure 1.

3.2 Reflectance sensitivity to aerosol type and cloud parameters

The sensitivity of the satellite measured reflectance at 354 and 388 nm to cloud-top height was first examined in the no-aerosol case. For a cloud of optical depth 10,

systematically varying cloud top level between 950 and 100 mb produced maximum differences of -3% reflectance at 354 nm and -1% at 388 nm resulting from the shielding by clouds of molecular scattering. Negligible reflectance change was observed associated with variations in cloud drop effective radius.

Figure 2 shows the calculated top of the atmosphere reflectance at 388 nm as a function of cloud optical depth (COD) for a column atmosphere containing cloud layer between 1.0 and 1.5 km and an aerosol layer with peak particle concentration at 3 km above the surface and a full-width at half maximum of 0.5 km. Results for a no-aerosol case and two different absorbing aerosol models of optical depth 1 (at 388 nm) are presented.

The solid line in Figure 2 depicts the relationship between the calculated TOA 388 nm reflectance, and cloud optical depth for COD between 0 and 50. The observed reflectance increases monotonically with COD as expected. The cloud diffuse radiation is by far the most important source of the observed TOA reflectance enhancement. Although molecular and aerosol scattering are also present [Wen *et al.*, 2008], their contribution to the observed TOA reflectance for overcast conditions, which may be important for optically thin clouds, is negligible at large values of cloud optical depth.

The results associated with an aerosol layer of single scattering albedo of 0.95 (at 388 nm) representative of BC are indicated by the dotted line in Figure 2. For weakly absorbing aerosols, scattering dominates over absorption effects and the net result is an increase of TOA reflectance values above those of the cloud-only case for optically thin clouds. Aerosol scattering and absorption effects cancel each other at a COD value of about 5. At larger COD values the increased cloud reflectance triggers additional aerosol

absorption, resulting in TOA reflectance values lower than the ones for the no-aerosol case. For the largest modeled COD (50) the weakly absorbing aerosol produces a 16% TOA decrease in relation to the no-aerosol case. For a stronger absorbing aerosol case (SSA=0.85) the absorption effect dominates over scattering even at low COD values as indicated by the dashed line in Figure 2. For a COD value of 50, the observed TOA reflectance is reduced by 33%.

3.3 Aerosol Index sensitivity to spectral aerosol absorption.

The resulting AAI for clouds only and above-clouds absorbing aerosol layers are shown in Figure 3. For clouds only the AAI is slightly negative, reaching a value of -0.6 at COD 5. It then increases with increasing COD, becoming zero at COD 20 and reaching about 0.25 at COD 50. Although the AAI of clouds depends on observing geometry (solar and viewing zenith angles, and relative azimuth angle), for solar zenith angles lower than 60, it oscillates around zero between -1.0 and 1.0 (not shown) with the slightly negative values associated with COD smaller than about 20 and small positive values for larger COD's. These results shown in Figure 3 are consistent with previously reported sensitivity analysis using Mie Theory [Torres *et al.*, 1998; Herman *et al.*, 1997] that show the weak spectral dependence associated with the scattering effects of non-absorbing large size particles. For typical AOD values, large aerosol particles yield AAI's close to zero whereas for non-absorbing aerosol particles smaller than about 0.3 μm and typical AOD values the resulting AAI is negative reaching values as low as about -1 for large aerosol loads. The slightly larger cloud-only AAI values in Figure 3 than reported by Torres *et al* [1998] are due to the much larger optical depth of clouds.

299 The small AAI values calculated for cloud droplets in the current work differ
300 from recently published calculations [*Penning de Vries et al*, 2009] in which clouds are
301 reported to produce negative AAI values (as large as -1.5) for all values of COD. A
302 likely explanation for the noted discrepancy is the *Penning de Vries et al.* approximate
303 representation of the angular dependence of scattering by clouds and aerosols using a
304 Henyey-Greenstain (H-G) phase function instead of actual Mie calculations. Using the
305 H-G function to represent clouds does not allow to account for the angular effects
306 associated with varying size of the scattering particle [*Hansen*, 1969].

307 For absorbing aerosols located above clouds, the radiative coupling between
308 cloud diffuse radiation, molecular scattering, and particle absorption enhances the
309 observed near UV spectral contrast and, therefore, the resulting AAI is larger than the one
310 associated with the same aerosol layer under cloud-free conditions. The AAI magnitude
311 will also depend on the spectral dependence of aerosol absorption as determined by the
312 wavelength dependence of the imaginary component of the index of refraction. The
313 effect of the above-cloud absorbing aerosols on the calculated AAI as a function of COD
314 for several values of the Angstrom Absorption Exponent (AAE) [*Bond*, 2001] and SSA
315 0.85 (388 nm) is also shown in Figure 3. In all cases the calculated positive AAI is a
316 strong function of the COD of the underlying cloud layer. The slope of the observed
317 COD-AAI relationship decreases for COD values larger than 20. The resulting AAI
318 values increase rapidly as the AAE varies from a value of 1.0 (dotted line), an indicator
319 of non-wavelength dependent imaginary refractive index (representative of BC), to
320 values of 2.0 (dot-dash line), and 2.8 (dashed line) representative of wavelength-

dependent aerosol absorption. A detailed analysis of the AAI sensitivity to spectral aerosol absorption is presented in *Jethva and Torres* [2011].

The presence of organic carbon (OC) in the carbonaceous aerosols produced by biomass burning has been demonstrated by a variety of laboratory and field studies [Kirchstetter et al., 2004; Hoffer et al, 2006; Russell et al., 2010]. A recent analysis combining OMI near UV observations, radiative transfer calculations and ground-based measurements of aerosol optical depth [Jethva and Torres, 2011] has further confirmed the presence of OC as an important aerosol component of biomass-burning generated smoke. *Jethva and Torres* [2011] showed that allowing for the wavelength dependence of the imaginary refractive index in the near-UV (an indication of OC presence) produced significantly better agreement of OMI retrieved AOD with sun-photometer observations than when using an aerosol model with wavelength independent imaginary refractive index characteristic of black carbon. Thus, the OC aerosol model has been selected as more representative of the actual carbonaceous aerosol load in the OMAERUV algorithm. In the remainder of this work we will use the OC aerosol model.

3.4 Aerosol Index Sensitivity to Aerosol-cloud separation

The AAI dependence on aerosol layer height over cloudless scenes is a well documented AAI property [Torres et al, 1998; Degraaf et al, 2005]. We will now examine the resulting height dependence when the absorbing aerosol layer lies above a cloud deck of varying reflectance as determined by the COD. As in the preceding analysis, the cloud layer is located between 1.0 and 1.5 km. The height of the absorbing aerosol layer is assumed to be 2.5, 3.0, 4.0 and 5.0 km above the surface. The resulting relative change in AAI value as a function of aerosol layer height and COD is depicted in

Figure 4. Largest percent change is observed for the cloud-free case ($COD = 0$) and quickly goes down with increasing COD. For a COD value of 30, the AAI associated with a height change of 2.5 km is about 25 %, compared to more than 75% for the cloud-free case. Thus, unlike in the cloud-free case where the aerosol layer height is a large source of uncertainty for the quantitative interpretation of the AAI [Torres *et al.*, 1998], in the presence of underlying clouds the AAI sensitivity to aerosol layer height is reduced. The inset in figure 4b illustrates the rate of change of AAI per 1km change in aerosol layer height as a function of cloud optical depth. The observed rapid drop in AAI sensitivity to cloud-aerosol separation with increasing COD is an advantage from retrieval point of view as it introduces less uncertainty in the derivation of AOT due to wrong assumption on the aerosol height above the cloud.

4. Retrieval Approach

4.1 Simultaneous aerosol-cloud retrieval

It the preceding sections it has been shown that the magnitude of the observed AAI associated with aerosol layers above clouds depends on the reflectance of the underlying cloud (COD), the aerosol load (AOD) and single scattering albedo, the AAE parameter, and the aerosol-cloud separation. For testing of the current research retrieval algorithm the values of some of these parameters can be prescribed based on independent observations. Aerosol-cloud separation, for instance, is obtainable from space-borne CALIOP lidar profiles. A justifiable assumption of the 388 nm single scattering albedo can be made from retrievals by the cloud-free OMAERUV algorithm [Torres *et al.*, 2007] in the vicinity of the cloud-aerosol feature under investigation assuming this value

to be representative of the SSA of the above-cloud aerosol load. Assumption on the value of the AAE parameter is required. In principle, COD information from other sensors in the A-train (i.e., MODIS) could be used as input to the inversion algorithm. This option, however, implies the use of erroneous COD data as the MODIS cloud optical product will be most probably also affected by the absorption effects of the overlying aerosol layer [Wilcox *et al.*, 2009]. In this work we demonstrate a retrieval approach that accounts for the spatial variability of clouds and aerosols by simultaneously deriving AOD and COD over overcast scenes covered by opaque clouds.

The retrieval scheme uses a set of pre-calculated values of AAI and 388 nm reflectances for several values of COD (0, 2, 5, 10, 20, 30, 40, 50), and AOD (0, 0.1, 0.5, 1.0, 2.5, 4.0) for a fixed value of SSA and assumed AAE. Cloud and aerosol layer heights are prescribed based on CALIPSO observations. With the above stated assumptions the interpolating domain reduces to a two dimensional array on COD and AOD as illustrated in Figure 5. As shown on the web-like diagram, an observation set of AAI and the 388 nm reflectance is associated with a set of values of COD and AOD.

4.2 Error Analysis

The accuracy of the retrieved parameters depends on the uncertainty associated with the assumed values of aerosol layer height above the cloud and the wavelength-dependent aerosol single scattering albedo. In this section we discuss the results of an error analysis assuming a set of ‘true’ conditions consisting of an aerosol layer of known optical depth and single scattering albedo located at a height Z above the surface, and a 0.5 km thick cloud layer with cloud top at 1.5 km and known optical depth. The assumed

cloud-aerosol vertical structure is representative of the typically observed cloud-aerosol layers over the Southern Atlantic-Ocean off the coasts of Angola and Namibia during that region's biomass burning season [King *et al.*, 2003; Chand *et al.*, 2009]. The uncertainty on the spectrally dependent single scattering albedo is evaluated by assuming a fixed SSA value at 388nm and an associated AAE value that describes the wavelength dependence of the aerosol absorption optical depth. The true value of Z is assumed to be 4km. A SSA value of 0.85 typical of biomass burning smoke aerosols for this region [Eck *et al.*, 2003], and an AAE value of 2.2 as reported by Kirchstetter *et al.*, [2004] during SAFARI 2000 field campaign are assumed. Radiative transfer calculations were carried out for a nominal AOD value of 0.5, and COD values of 5 and 10. The assumed values of Z and SSA are perturbed by ± 2 km and ± 0.03 respectively that represent typical variability associated with these parameters [Chand, *et al* 2009; Eck *et al.*, 2003] over the Southern Atlantic region. The assumed AAE parameter is varied between 1.8 and 2.6 or ± 0.4 to account for possible variations in aerosol composition (i.e., OC content). Although the actual natural variability of AAE is largely unknown the tested range allows accounting for some uncertainty on its actual value. The AAE perturbation is equivalent to a change from the nominal spectral dependence of 15% in the values of the imaginary refractive index (i.e., $k_{354} = 1.15k_{388}$) to 10% and 20%.

The obtained AOD and COD retrieval errors associated with the assumed range of uncertainty of the prescribed parameters are shown in Table 2. For a nominal COD value of 5, maximum AOD retrieval errors take place when Z is underestimated (40%), SSA is overestimated (48%), and AAE is underestimated (23%) yielding a combined largest AOD error estimate of 67%. Minimum AOD errors, on the other hand, result

when Z and AAE are overprescribed and SSA is underestimated producing a combined AOD under-estimation error of about -34%. The overall AOD error goes down with increasing cloud optical depth. For a more typical COD value of 10 the overall AOD error range decreases to -27 to 54%.

Expected errors in retrieved COD are smaller than those associated with the AOD retrieval. For a nominal COD value of 5 the overall error range associated with the above described uncertainties -4 to 7%, and gets larger as the cloud becomes optically thicker. An error range between -6 to 17% is expected for COD 10.

5. Case studies

5.1 Cloud-only case: October 13, 2006

We will first examine the near-UV capability of retrieving COD from single channel-measurements. This test is necessary to assess the results of the combined AOD-COD retrieval algorithm discussed in this section. In the absence of intervening aerosols cloud optical depth can be derived from solar reflectance measurements at wavelengths where gas absorption interference is negligible. Cloud optical depth is routinely derived from MODIS observations on the Terra and Aqua satellites at 860 nm over the oceans and 645 nm over land [Platnick *et al.*, 2003]. In the cloud-only retrieval application the observations are inverted using the forward calculations for AOD zero shown in Figure 5. This is equivalent to a single channel retrieval using OMI's 388 nm reflectance measurements. This approach was used to derive the optical depth of the cloud field over the Southern Atlantic Ocean on October 13, 2006. To ensure that broken clouds were excluded, the retrieval was only applied to those OMI scenes characterized by overcast

conditions or unity MODIS geometric cloud fraction (CF). The absence of absorbing aerosols above the clouds was verified by making sure the AAI was close to zero. A comparative analysis of MODIS geometric CF and OMI 388 nm reflectivity (introduced in section 2). indicated that overcast conditions can be present at OMI reflectivity (ρ) values as low as 0.20. Thus, a 0.20 minimum ρ threshold value was used. OMI retrieved COD was compared with collocated MODIS COD observations. Given the large difference in spatial resolution between the two sensors (MODIS $0.5 \times 0.5 \text{ km}^2$ versus OMI $13 \times 24 \text{ km}^2$) linear averaging of the MODIS COD product over the OMI pixel was necessary. Data points in Figure 6 are color-coded as a function of the associated OMI derived reflectivity. The comparison of the OMI research COD (388 nm) and MODIS operational COD (860 nm) products in Figure 6 indicates a close agreement between the retrievals with correlation coefficient 0.92 and slope 0.96. The OMI COD values appear slightly lower than those from MODIS for R values lower than 0.30 probably associated with the presence of broken and/or transparent clouds. The close OMI-MODIS agreement in retrieved COD illustrates the sensitivity of OMI observations to cloud presence and is consistent with the reported agreement in effective CF calculated by *Stammes et al* [2008] from MODIS retrieved geometric CF [*Platnick et al.*, 2003] and OMI retrieved effective CF from observations of $\text{O}_2\text{-O}_2$ absorption at 477 nm [*Acarreta et al.*, 2004].

5.2 Aerosols above clouds: August 4, 2007

The presence of smoke layers above stratocumulus clouds off the coasts of Namibia and Angola is a common occurrence during the biomass burning season from August thru October. Low level stratus clouds form a few hundred meters above sea

level. Since the continental surface is about 1 km above sea level, westward smoke advection results in the accumulation of carbonaceous aerosols above low level clouds. The retrieval method described in section 4 has been applied to the aerosol event depicted in Figure 1. Note that during this event a section of the aerosol strip detected by CALIOP is located above a thin, spatially continuous cloud deck at 1 km above sea level (between about 21°S and 11°S) whereas cloudy and cloud-free conditions are intermittently present below the aerosol layer between 11°S and 5°S. Cloud presence above the aerosol layer is observed north of 5°S. For retrieval purposes, CALIOP observations shown in Figure 1 have been used to prescribe cloud location (top 1.5 km, bottom 1 km) and aerosol layer height (3km). A 388 nm aerosol SSA value of 0.88 has been assumed based on OMAERUV algorithm retrieval [Torres *et al.*, 2007] over a cloud free region at 12°S, 10°E. The spectral dependence of near-UV aerosol absorption has been characterized based on the work by Kirchstetter *et al* [2004] who found that an AAE value of about 2.2 adequately described the spectral dependence of aerosol absorption optical depth of carbonaceous particles resulting from biomass burning during the SAFARI-2000 campaign [Swap *et al*, 2003]. An AAE value of -2.2 is equivalent to a 14% increase in imaginary refractive index from 388 to 354 nm as assumed in this analysis. Retrievals were carried out for OMI scenes with R larger than 0.20 and AAI values larger than unity. Note that in the presence of absorbing aerosols above clouds, the actual cloud reflectivity is likely larger than the OMI observed one.

Figure 7 depicts the retrieved AOD (Figure 7a) and COD (Figure 7b) fields. The number of possible retrievals is limited by the upper limit COD limit of 50 currently used in the research retrieval algorithm. Therefore for COD's larger than 50, neither COD nor

AOD values are reported. A clear south-north gradient in the obtained AOD is observed across the aerosol layer with minimum values in the 0.3-0.6 range along the southern edge of the layer and maxima of 1.8 or larger along the northern boundary of the aerosol layer. The COD field shows three localized areas of COD larger than about 12 whereas the COD background values of about 5 to 10 predominate everywhere else.

A qualitative assessment of the derived aerosol optical depth can be done by examining certain characteristics of the retrieved AOD and COD fields to obtain an idea of the reliability of the results or the lack thereof. A quick visual inspection of the spatial variability of the two retrieved quantities indicates that the derived AOD and COD fields are not generally correlated. For instance, low AOD and high COD values are observed at the region in the vicinity of 14°S, 2°E, whereas the opposite scenario (i.e., high AOD and low COD values) occurs in the area surrounding the clear area at 12°S, 7°E.

Since Aqua-MODIS and Aura-OMI observations are only a few minutes apart, one can evaluate the OMI COD retrieval by directly comparing it to MODIS results. As it was previously demonstrated in this work, when no aerosols are found above clouds OMI retrieved COD is in excellent agreement with the MODIS operational COD product (Figure 6). In spite of the difference in spatial resolution between the two sensors the different cloud features in the MODIS product are easily recognizable in the OMI COD image (not shown). A scatter plot of retrieved COD by the two sensors is shown in Figure 8. Because of the aerosol absorption effect on the MODIS 860 nm reflectance [Wilcox *et al.*, 2009], OMI retrieved COD values are expected to be higher than those of MODIS. Overall a good level of agreement between the two retrievals is observed as indicated by the high correlation coefficient (0.85), near-one slope (1.07) and low y-intercept (-

0.19). A careful analysis in terms of cloud reflectivity shows that for ρ values less than 0.25 (blue filled circles) OMI consistently retrieves lower-than-MODIS COD 's. This result probably indicates that OMI clouds of reflectivity less 0.25 are either non-opaque or non-uniform (i.e., broken clouds), or both, which would allow radiation from below the cloud or from cloud-free parts of the pixel reach the sensor producing an underestimate of the cloud optical depth. For clouds with reflectivity between 0.25 and 0.30 (green triangles) OMI and MODIS COD tend to be in agreement for COD values up to about 7. At larger values the OMI COD is again underestimated in relation to MODIS probably due to no meeting the required overcast conditions. Thus, it seems that for ρ values less than 0.30, the overcast and opaque cloud condition is not met and, therefore, the OMI COD error is comparable to or larger than the aerosol absorption induced error in the MODIS product. For ρ values larger than 0.30 (red crosses), on the other hand, OMI consistently retrieves higher than MODIS values for COD values up to 15. The lower MODIS values are most probably associated with the aerosol absorption effects at 860 nm that would result in the underestimation of the optical depth as discussed by *Wilcox et al* [2009]. Figure 9 shows a scatter plot between AOD and AAI, indicating the expected high correlation between these two quantities.

Another way of qualitatively assessing the retrieved AOD fields is to analyze the spatial continuity between cloudy and cloud-free areas. An inherent assumption made here is that the AOD field does not change significantly from overcast scene to the adjacent clear region. Here, we employ standard OMAERUV algorithm to derive AOD fields over cloud-free region. Figure 10 shows a composite of AOD retrievals over cloudy and cloud-free scene. No obvious spatial discontinuities in AOD along the

northern edge of cloudy region and adjacent clear region are observed. Missing data, however, is observed along the northernmost edge of the cloud and, especially over the southernmost boundary in the transition from overcast to totally clear conditions. The occurrence of these pockets of missing data is expected as none of the OMI algorithms deal with partial cloudiness conditions.

5.3 Aerosols above clouds: August 31, 2005

A second sample retrieval was carried out for an aerosol event on August 31, 2005 over the same region. Similar assumptions on aerosol-cloud vertical distribution to those in the August 4-2007 case were used for this event. A 388nm SSA value of 0.90 retrieved over a clear area at 16°S, 14°E was assumed together with an AAE exponent of -2.2 as in the previously discussed example.

The MODIS true color image composite and the OMI AAI data on August 31 2005 (not shown) illustrate the presence of a smoke layer located above a large horizontally extended cloud deck over the Southern Atlantic Ocean. Retrieved AOD and COD fields for this event are depicted in Figure 11. The shown AOD field includes retrievals under both overcast and cloud free conditions. Peak AOD values larger than 3.0 are observed over the southeastern region of the cloud off the coast of Namibia where cloud optical depth values in the vicinity of 5 were retrieved. Lower AOD values of about 1.0 are observed over the optically densest part of the cloud where the COD reaches values close to 20. As in the previous case no correlation between the retrieved AOD and COD fields is observed. The OMI retrieved COD again correlates very well with the MODIS COD product as shown in Figure 12. As in the previous case, the OMI-

derived COD is generally higher than the MODIS retrieval for reflectivity larger than 0.30 and, somewhat lower than the MODIS results for lower reflectivity values. In this case, larger OMI-MODIS differences are observed than on the August 4, 2007 case which is consistent with the retrieved larger aerosol load.

5.4 Assessment of AOD results

The validation of satellite retrievals of aerosol optical depth is only possible by means of sun-photometer measurements as it is routinely done to validate space-borne quantification of the atmospheric aerosol load. For a direct quantitative assessment of above-cloud aerosol optical depth retrievals, as reported in this work, airborne sun-photometer measurements [Russell *et al.*, 2005] would be required. In the absence of those observations, however, a semi-quantitative evaluation can be attempted by comparing the OMI obtained AOD results to those obtained by other satellite-based techniques capable of detecting, and possibly quantifying, aerosol amounts above clouds. Such retrievals have been attempted by application of active [Kittaka *et al.*, 2011; Chand *et al.*, 2008] CALIOP observations as well as passive POLDER measurement of polarized radiation [Waquet *et al.*, 2008] described in section 2.

A comparison of the CALIOP version 3.01 standard AOD (532 nm) to OMI AOD at 388 nm for the August 4 2007 case discussed in section 5.2 was carried out. Although a correlation between the two retrievals is apparent, the standard CALIOP AOD is significantly smaller than the OMI results. The wavelength difference does not explain the large observed differences. Given the current provisional nature of the CALIOP AOD product as indicated by the official CALIPSO Quality Statement, we defer further evaluation of OMI above-cloud-aerosol retrievals using CALIOP data as a future work

activity when a more definitive CALIOP AOD product may be available. Results of above-cloud-aerosol optical depth by the method described here were compared to an equivalent POLDER AOD product as described by [Waquet *et al*, 2008]. Figure 13 shows a scatter plot of POLDER AOD at 865 nm to OMI's 388 nm for the event of August 4 2008 documented in Waquet *et al* [2008]. A well defined correlation between the two measurements is observed. Although OMI retrieved AOD's are about three times as large as POLDER's, the difference can be explained by the small aerosol particle size typical of carbonaceous aerosols and the difference in reporting wavelength. A reduction of results to a common wavelength (not shown) indicates a close one-to-one agreement.

6.0 Summary and conclusions

We have discussed the basis, and presented examples, of an inversion procedure to derive the optical depth of the atmospheric aerosol load above clouds. The retrieval technique uses measurements of backscattered near-UV measurements by the OMI sensor on the Aura satellite. The OMI-derived absorbing aerosol index and the observed 388 nm reflectance are fed to an inversion procedure that simultaneously retrieves both aerosol and cloud optical depth. Results of a sensitivity analysis indicate that the magnitude of the aerosol index associated with absorbing aerosols above clouds depends on cloud and aerosol optical depth, wavelength dependent aerosol single scattering albedo (or AAE) and, to a lesser extent on aerosol layer-cloud separation. Thus, to simultaneously retrieve AOD and COD, assumptions on the values of the other parameters or external information on them is needed. An error analysis was carried out to estimate the uncertainty in the retrieved values of aerosol and cloud optical depth

associated with the uncertainty in the assumed values of single scattering albedo and aerosol cloud separation. Results indicate that the combined uncertainty of ± 0.03 in single scattering albedo and ± 2 km in aerosol-cloud separation yields an AOD error between -26% and 54% for typical cloud and aerosol layer optical depth of 10 and 0.5 respectively. Retrieval errors decrease with increasing cloud optical depth. Errors in retrieved COD are smaller than 20% in most cases which are comparable to the reported uncertainty of the MODIS product for optical depths larger than about 10 [Platnick *et al*, 2003].

The retrieval approach was applied to two aerosol events over the Southern Atlantic Ocean off the West coast of Southern Africa on August 4, 2007 and August 31 2005 where a persistent widespread layer of carbonaceous aerosols transported from the adjacent land region accumulates above a uniform low-level stratocumulus cloud deck during August and September. In the absence of airborne sun photometer measurements for a direct assessment of the validity of the retrieved AOD results, a qualitative analysis was carried out. For both events the OMI retrieved AOD and COD fields are uncorrelated with each other which indicate the absence of retrieval biases associated with the bright reflecting background. An examination of the spatial homogeneity of the AOD retrieval over overcast and cloud-free scenes August 4 2007 shows no obvious discontinuities in the clear-to-cloudy transition regions except for the expected lack of retrievals over partly cloudy scenes.

Direct comparisons of MODIS and OMI retrieved cloud optical depths during both events show a very good correlation. When the satellite derived scene reflectivity is lower than about 0.25 OMI retrieved COD's are smaller than those of MODIS indicating

the presence in the OMI scene of broken and/or transparent clouds. For scene reflectivities larger than 0.30, overcast opaque cloud conditions seem to exist and OMI COD retrievals are higher than those of MODIS. The larger OMI COD values are consistent with the expected error in MODIS COD associated with the effect of aerosol absorption on the near-IR radiances (860 nm) used in the retrieval. The good correlation between the two COD measurements and the explainable difference in OMI-MODIS retrieval results lends confidence to the results of the OMI two-parameter retrieval given that the OMI and MODIS COD retrieval approaches are fundamentally different from each other.

A quantitative evaluation of the derived AOD was carried out by comparing OMI AOD results to equivalent CALIOP and POLDER retrievals. The comparison to the lidar-based product was not conclusive due to the current provisional nature of the CALIOP AOD product. The comparison to POLDER results, on the other hand, yielded a very high correlation. The observed difference in magnitude of the two measurements can be explained in terms of the predominantly small aerosol particles and the difference in reporting wavelengths.

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810 Table 1.

Model	r_f (μm)	σ_f (μm)	N_f (cm^{-2})	r_c (μm)	σ_c (μm)	N_c (cm^{-2})	Refractive Index
BC aerosol	0.08	1.49	10.3	0.7	2.08	0.0021	1.50+0.015
OC aerosol	0.08	1.49	10.3	0.7	2.08	0.0021	1.50+0.015 (388 nm) 1.50+0.018 (354nm)

811 Aerosol parameters: mode radius(r), standard deviation(σ) and particle concentration(N)
812 for fine and coarse modes, and refractive index. The corresponding SSA is 0.85 for BC(
813 wavelength independent) and 0.85 and 0.84 at 388 and 354 nm respectively for the OC
814 model.

815 Table 2.

AOD	COD=5		COD=10	
	AOD	COD	AOD	COD
Z_{und} (2km)	40	4	26	9
Z_{ovr} (2km)	-19	-1	-12	-3
SSA_{und} (0.03)	-25	4	-23	1
SSA_{ovr} (0.03)	48	-5	43	-1
AAE_{und} (0.4)	23	3	19	14
AAE_{ovr} (0.4)	-14	-1	-6	-5

816 Percent error in retrieved AOD and COD for an aerosol layer height uncertainty of 1km.

817

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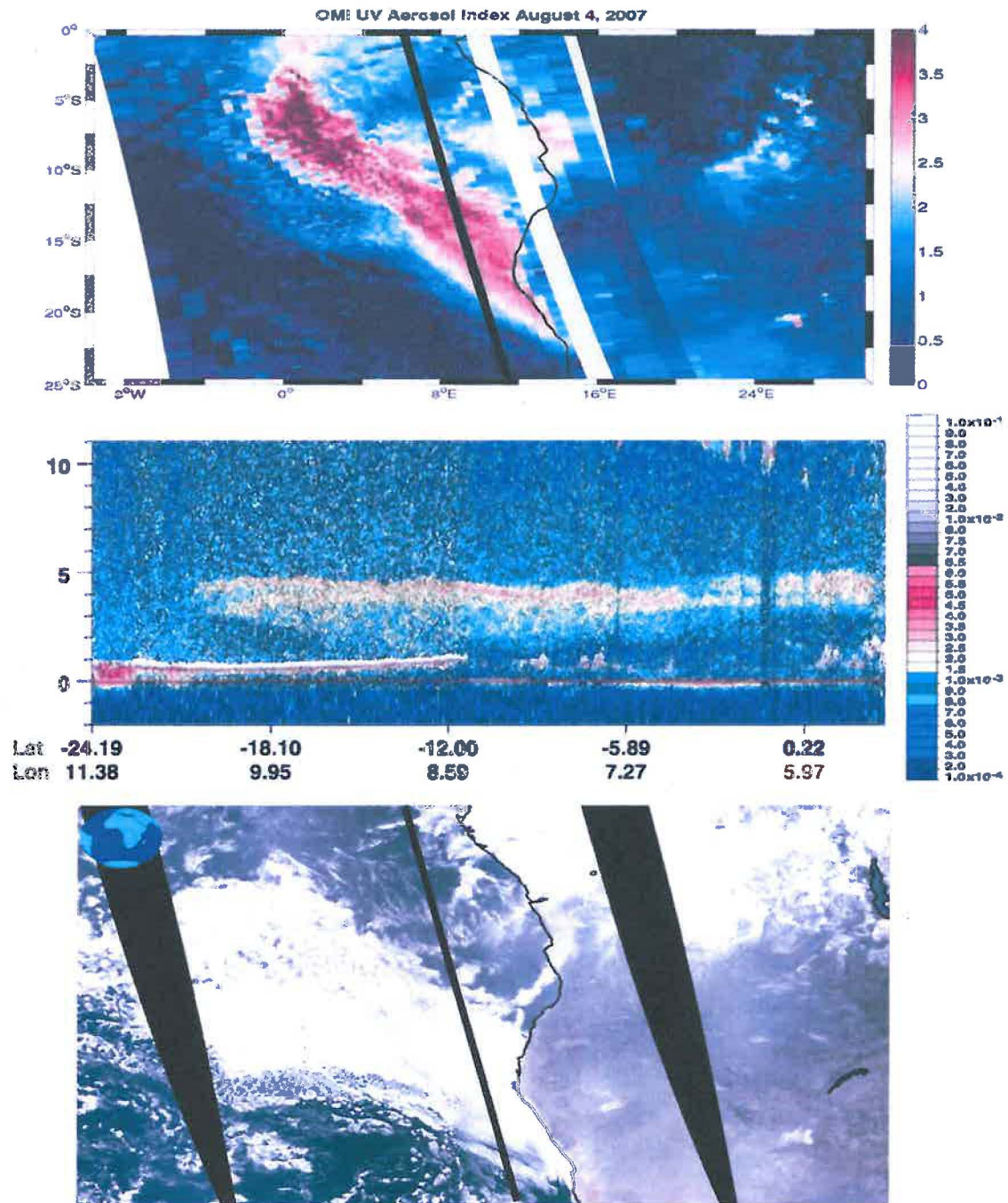


Figure1. OMI UV-AI (top panel), CALIOP back-scatter (middle panel), and MODIS true color image composite (bottom panel) for an aerosol event observed on August 4, 2007 off the coast of Central Africa and over South East Atlantic Ocean. The CALIOP track (thick black line) is also shown on UV-AI and RGB images.

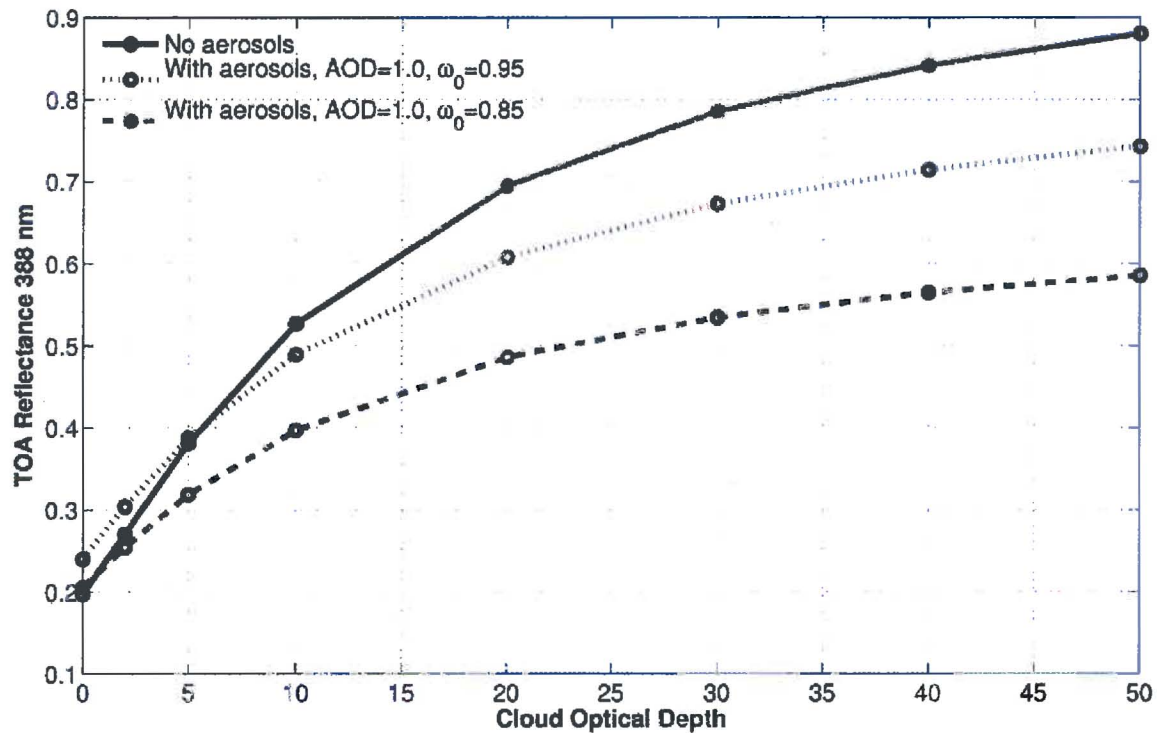


Figure 2. TOA reflectance sensitivity as a function of cloud optical depth for no-aerosol and for BC carbonaceous aerosol-laden atmosphere. The cloud layer is located between 1.0 and 1.5 km and the aerosol layer is at 3 km above the surface. Calculations were carried out for solar and satellite zenith angles 20° and 32° respectively, and relative azimuth angle 120° .

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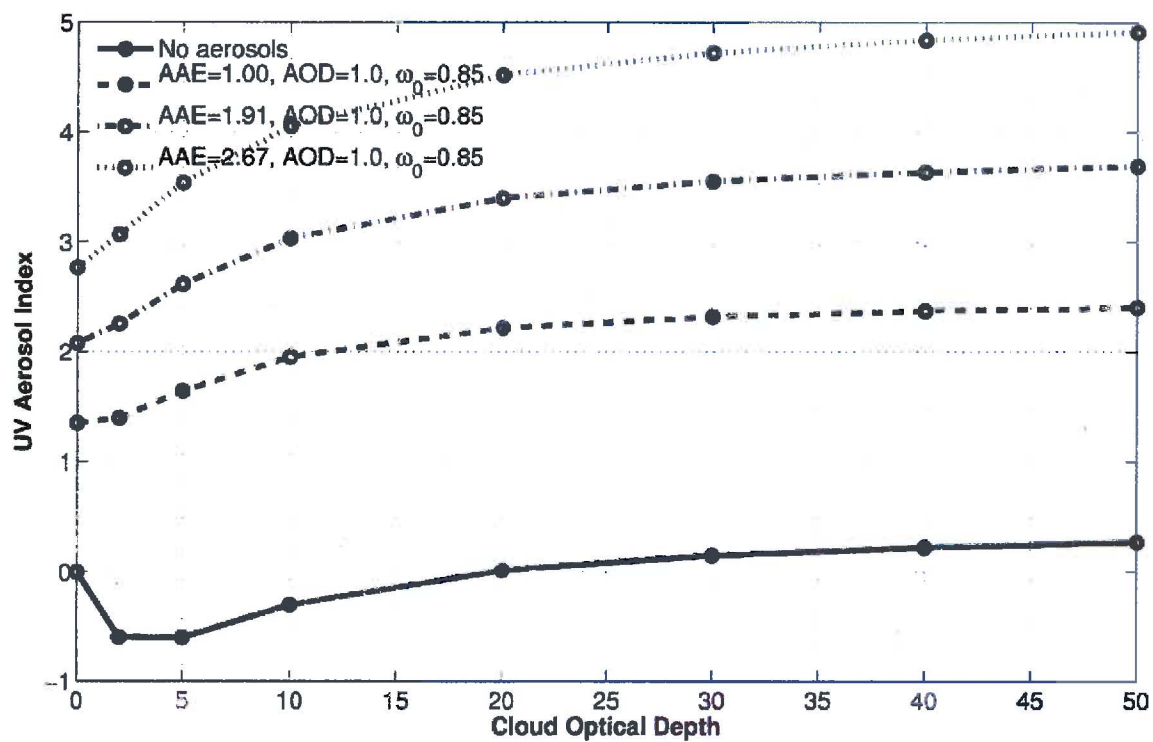


Figure 3. UV-aerosol index sensitivity to varying cloud optical depth for no aerosol, gray aerosol and colored aerosol models. Aerosol and cloud location and viewing geometry as in Figure 2.

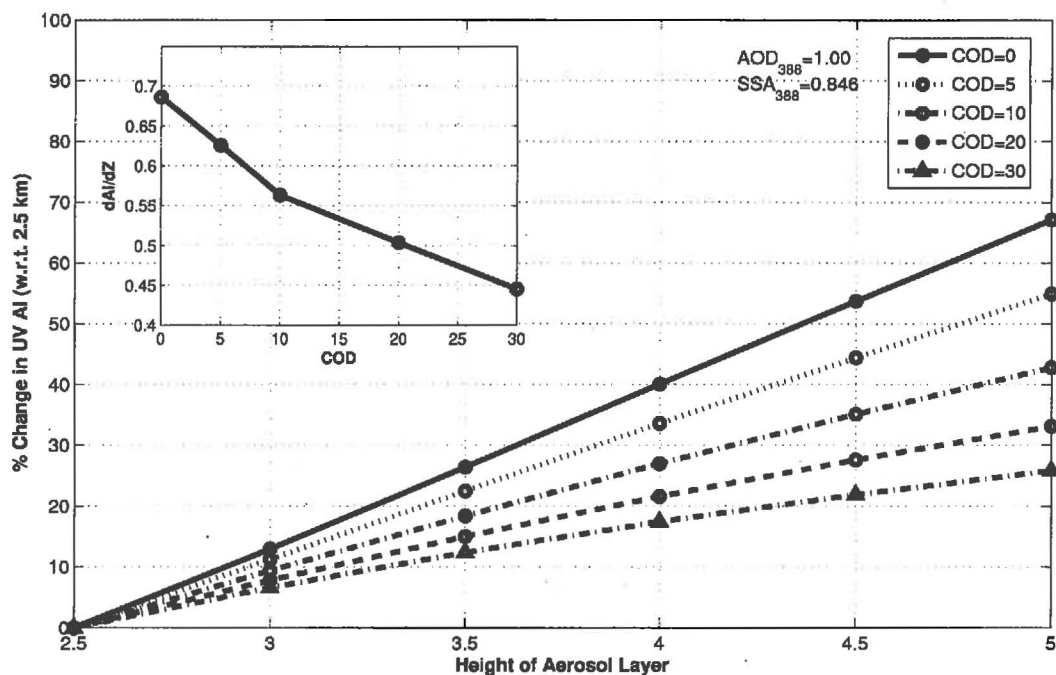


Figure 4. Relative change in UV-AI for varying aerosol mean height under different values of cloud optical depth. Shown in inset is the derivative of AI w.r.t. height as a function of cloud optical depth

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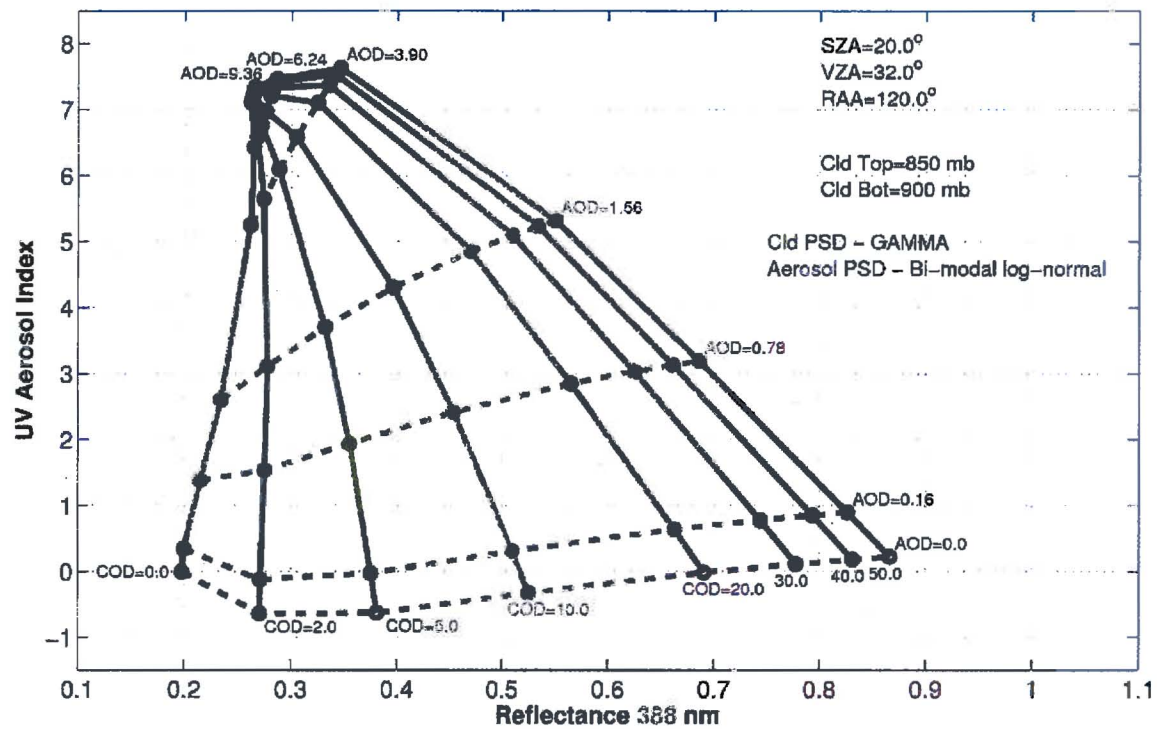


Figure 5. Retrieval domain of AAI Vs. Reflectance at 388 nm simulated using a radiative transfer model for several values of AOD at 388 nm (0.0, 0.156, 0.780, 1.56, 3.9, 6.24, 9.36) and COD (0.0, 2., 5., 10., 20., 30, 40, 50).

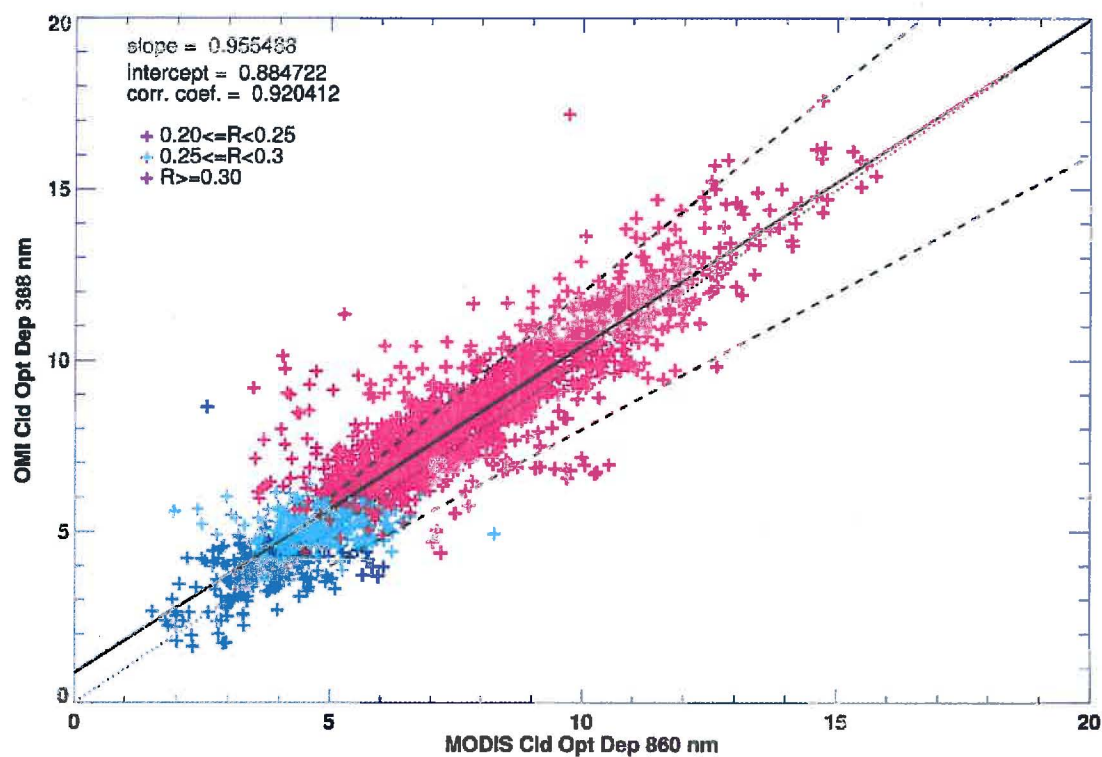
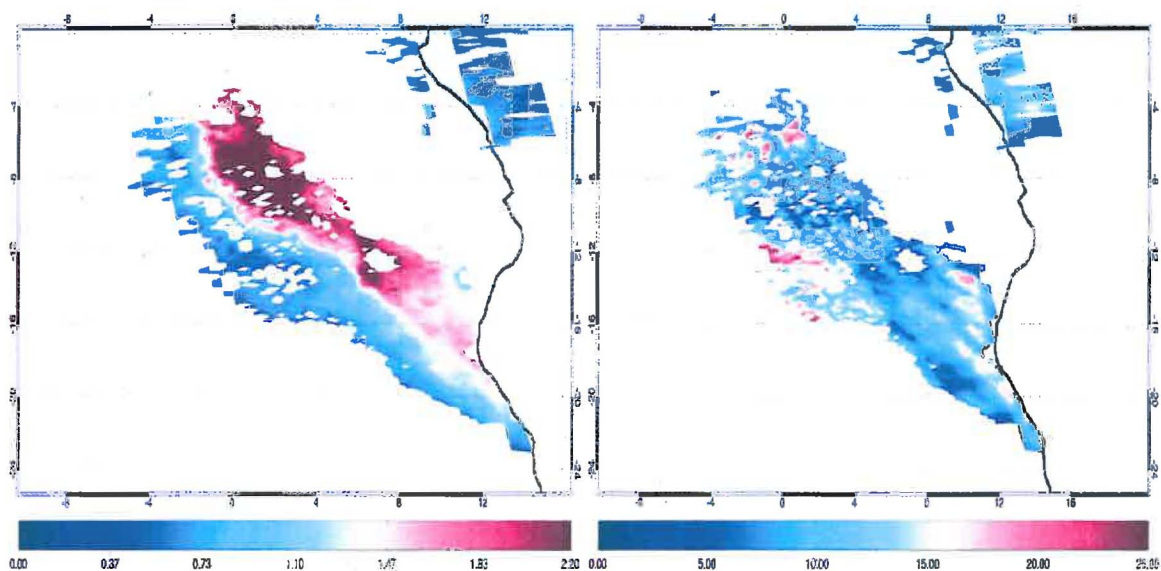


Figure 6. Comparison of MODIS and OMI cloud optical depth retrieved on October 13, 2006. No aerosols were present above clouds on this event. The dotted line indicates the one-to-one agreement, the solid line is the linear fit, and the dashed lines represent the $\pm 20\%$ uncertainty of the MODIS retrieval for COD's larger than 5. Significantly larger uncertainty exists for COD smaller than 5 [Platnick *et al.*, 2003].

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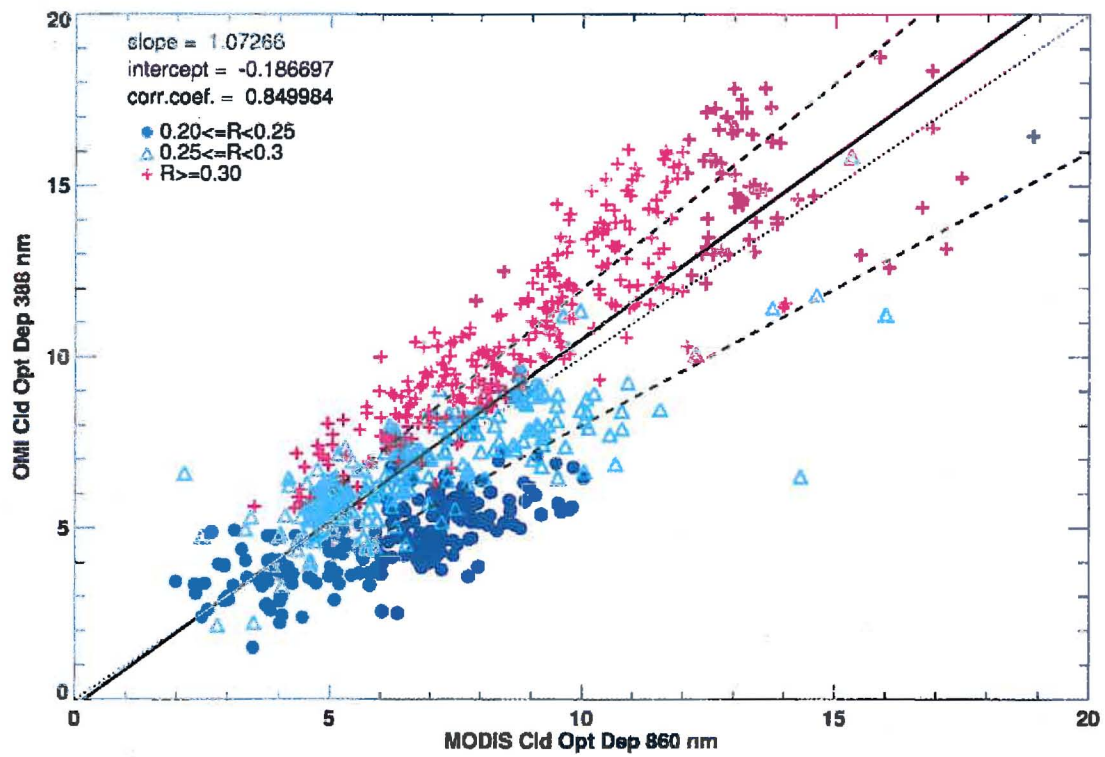
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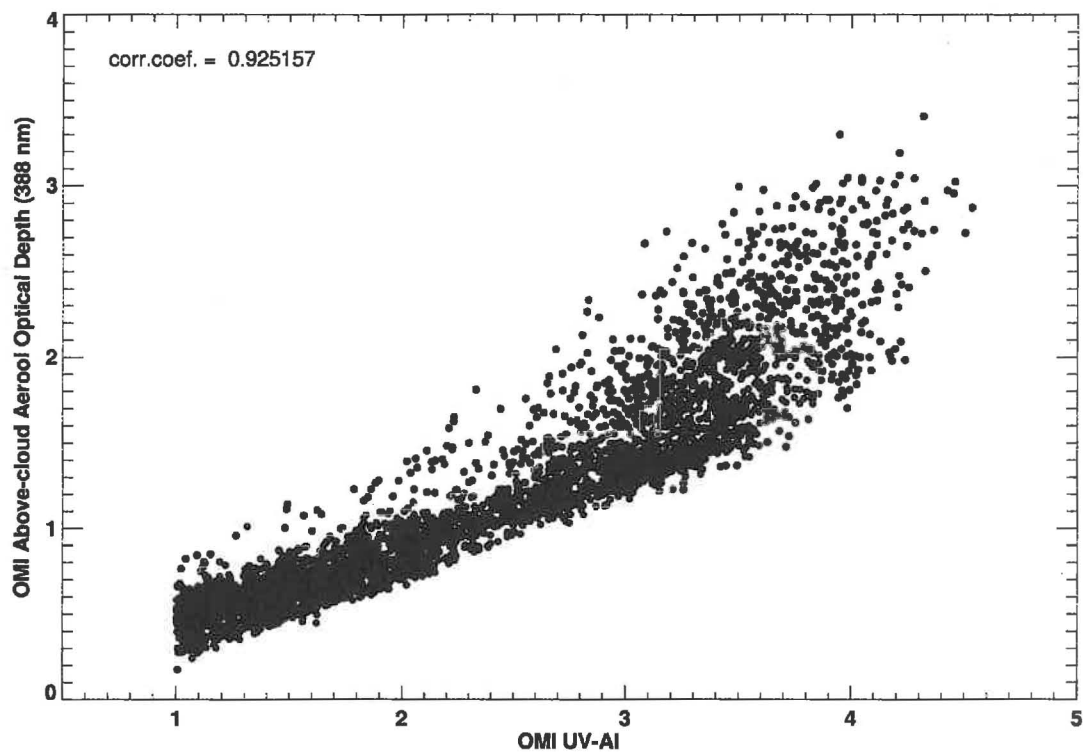
Figure 7. Spatial distribution of above-cloud AOD (388 nm) (left) and COD (right) for the Aug 4, 2007 aerosol event.

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Figure 8. As in Figure 6 for the aerosol event over the Southeast Atlantic Ocean on Aug 4, 2007.



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Figure 9. Relation between coincident aerosol optical depth and UV AI measured by OMI on Aug 4, 2007 over the study region.

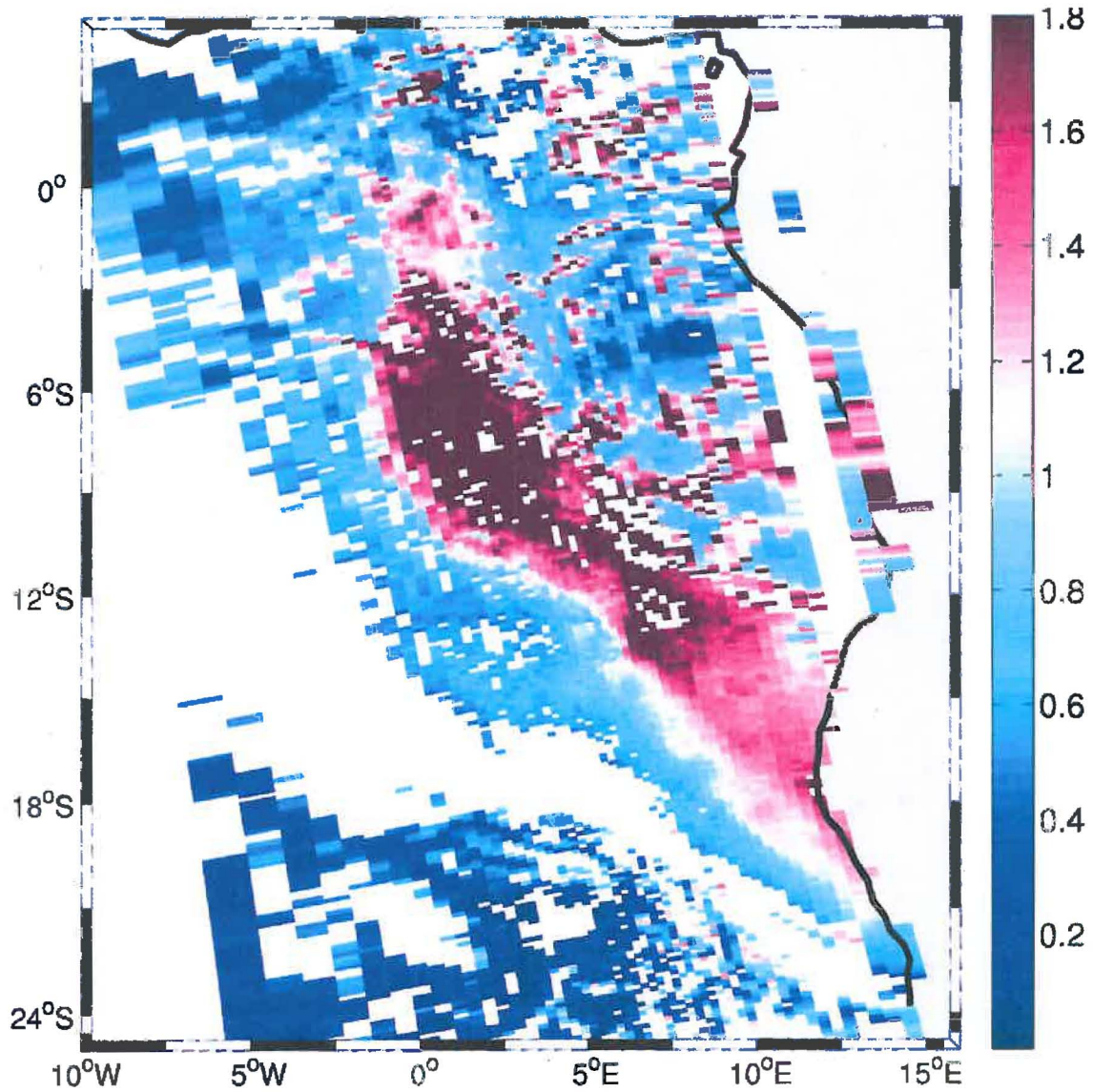
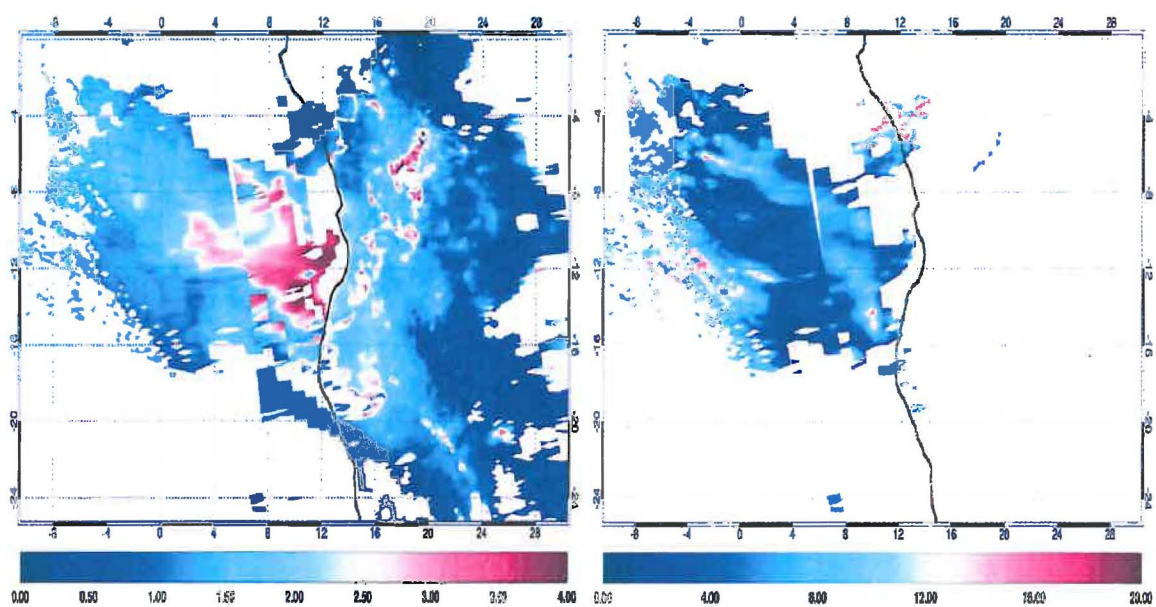


Figure 10. Composite of above-cloud and clear region AOD derived using cloud-aerosol and only aerosol OMAERUV algorithm on Aug 4, 2007.

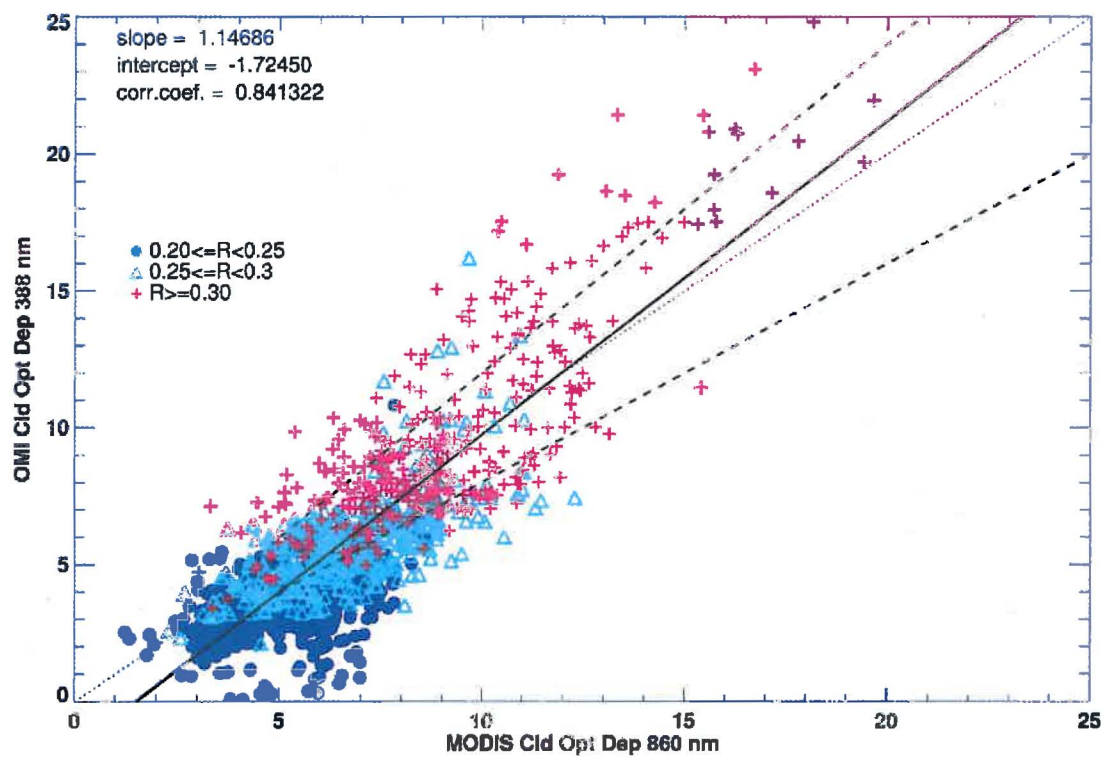
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Figure 11. Retrieved AOD (left) and COD (right) fields on August 31, 2005.

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Figure 12. As in Figure 6 for the aerosol event over the Southeast Atlantic Ocean on August 31, 2005

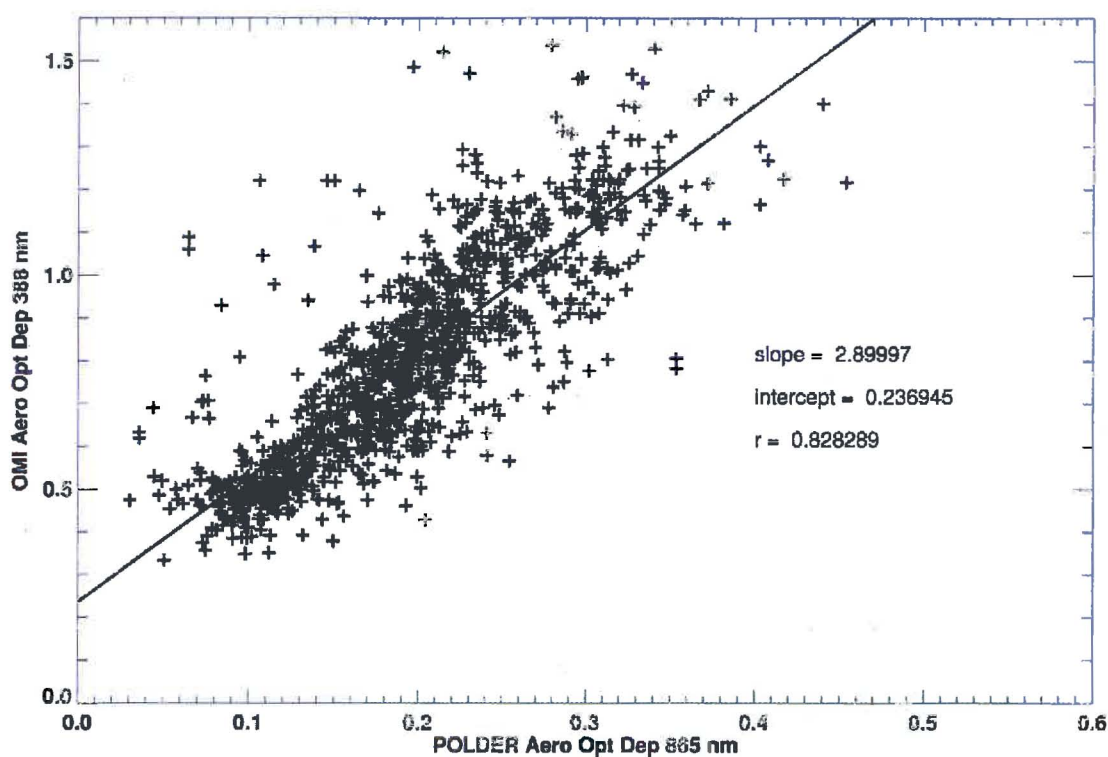


Figure 13. Scatter plot of retrieved 865 nm AOD from POLDER observation (x-axis) and 388 nm AOD from OMI measurements on August 4, 2008. The difference in magnitude is related to the small aerosols particle size and difference in reporting wavelengths (see text for details).